

# Disclosed New Evidence for Networking in Natural Rubber by Dielectric Relaxation Spectroscopy

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## Supporting Information

### Stress-strain curves of NR, DPNR and TE-DPNR

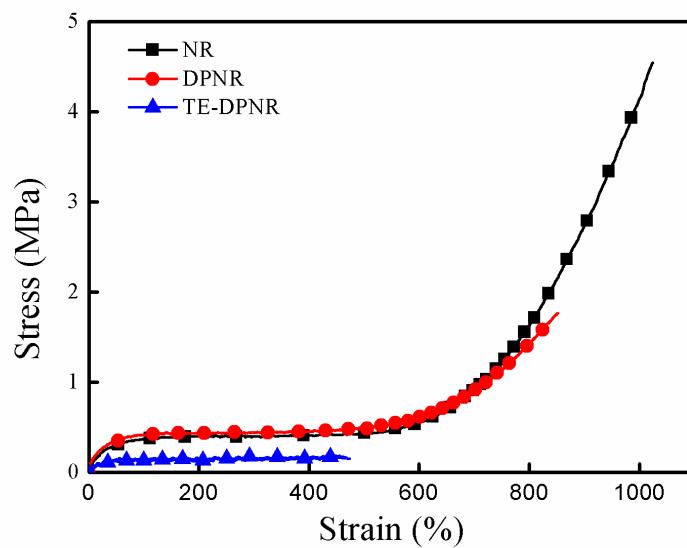


Fig. S1 Stress-strain curves of NR, DPNR and TE-DPNR

### SAXS curves of NR and DPNR

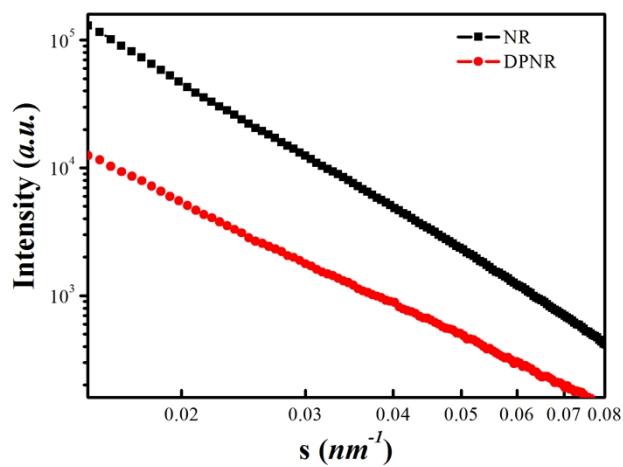


Fig. S2 SAXS curves of NR and DPNR

### $\epsilon'(T)$ of NR, DPNR and TE-DPNR

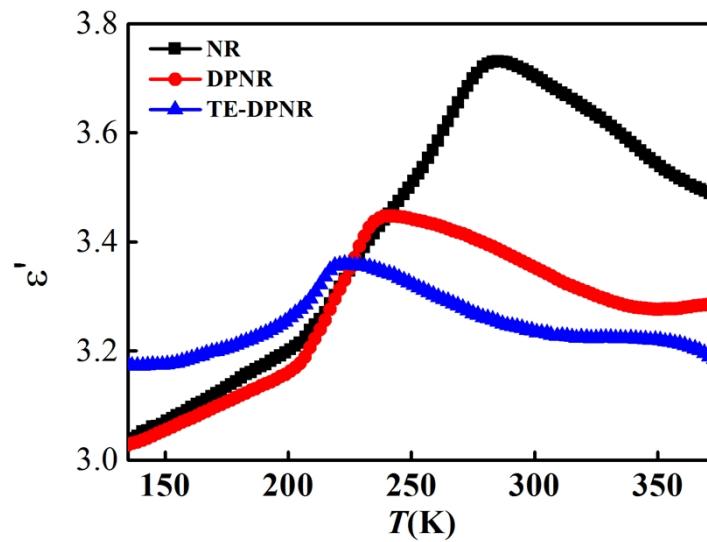


Fig. S3  $\epsilon'(T)$  of NR, DPNR and TE-DPNR at 7 Hz.

**Dielectric loss spectra of (a) NR and (b) DPNR (c) TE-DPNR using a HN function for fitting at 223K**

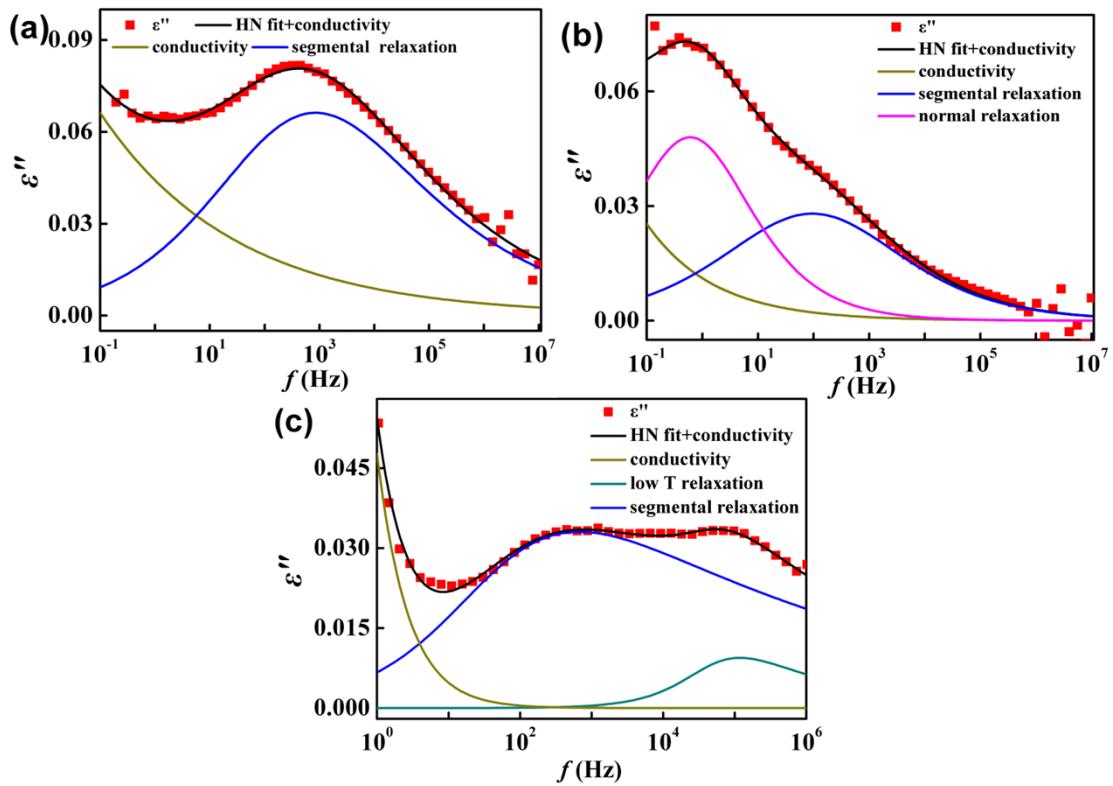


Fig. S4 Dielectric loss spectra of (a) NR and (b) DPNR (c) TE-DPNR using a HN function for fitting at 223K

### Dielectric loss spectra of NR using a HN function for fitting at 283K

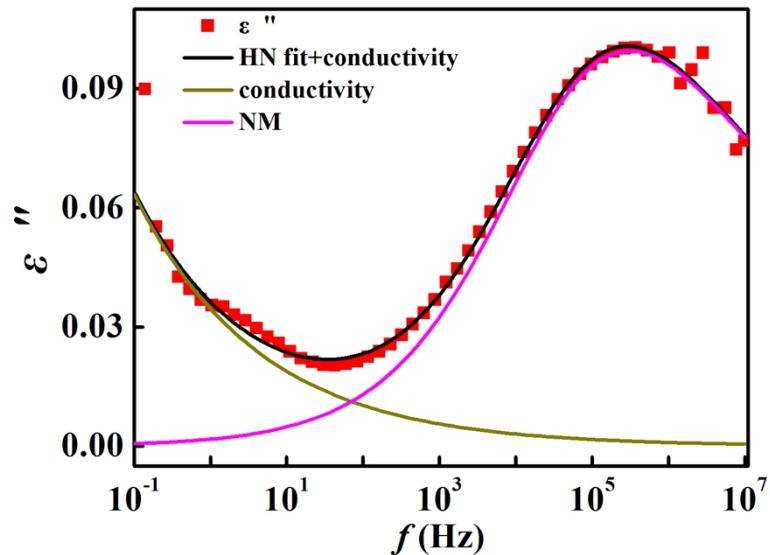


Fig. S5 Dielectric loss spectra of NR using a HN function for fitting at 283K

### Activation plot for SM, NM and the new mode relaxation for DPNR

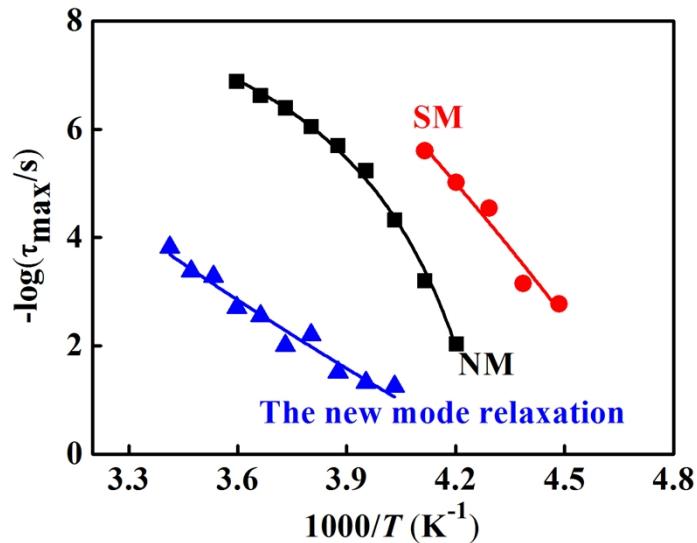


Fig.S6 Activation plot for SM, NM and the new mode relaxation for DPNR. The lines represent fits of Equation 6.

### Dielectric loss $\epsilon''$ vs. frequency of TE-DPNR

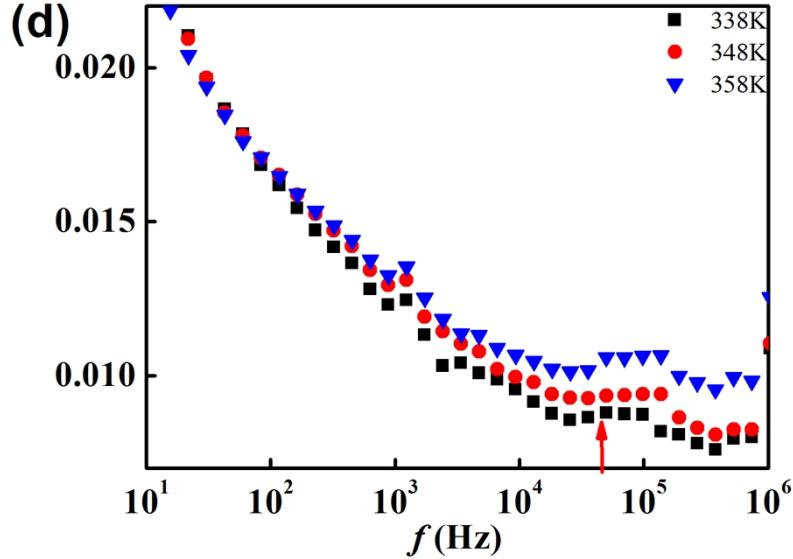


Fig.S7 Dielectric loss  $\epsilon''$  vs. frequency of TE-DPNR at 338K to 358K. The red arrows indicate the emerging of NM peaks at 338K

**Table S1** The peak value of  $\epsilon''$  for NM modes in Fig. 3.

Sample	Relaxation mode	Peak value of $\epsilon''$
NR	NM	0.0735
DPNR	NM	0.0523
TE-DPNR	NM	0.0196

**Table S2** Fitting parameters for SM and NM in the typical spectra illustrated in Fig. S4, Fig. S5, Fig. 6(b) and Fig. 8.

Sample	Mode	$T/K$	$\alpha$	$\beta$	$\tau_0/s$	$\tau_{max}/s$
NR	SM	223	0.3713	0.6208	$6.568 \times 10^{-4}$	$1.886 \times 10^{-4}$
DPNR	SM	223	0.3851	1	$1.675 \times 10^{-3}$	$1.675 \times 10^{-3}$
TE-DPNR	SM	223	0.5218	0.2061	$4.239 \times 10^{-3}$	$2.372 \times 10^{-4}$
NR	NM	283	0.4535	0.3442	$4.940 \times 10^{-6}$	$5.156 \times 10^{-7}$
DPNR	NM	283	0.7090	0.7585	$1.212 \times 10^{-7}$	$8.550 \times 10^{-8}$
DPNR	The new mode	283	0.4156	0.9740	$5.529 \times 10^{-4}$	$5.201 \times 10^{-4}$
TE-DPNR	LTM	173	0.3816	1	$3.944 \times 10^{-3}$	$3.944 \times 10^{-3}$